

Characterization of the effect of short pulse exposure on laser damage size, morphology, and conditioning in wide band gap materials

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Characterization of the effect of short pulse exposure on laser damage size, morphology, and conditioning in wide band gap materials

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Abstract

The first objective of this proposal was to develop a fundamental understanding of how short-pulse (100 ps to 1 ns) laser parameters affect conditioning and damage initiation in KDP and DKDP crystals. In this study we experimentally determine the effect of short-pulse laser parameters including pulse duration, pulse shape, and fluence on conditioning effectiveness, pinpoint damage density, pinpoint size, and pinpoint morphology in crystals. Based on the experimental results we developed empirical models, which give insight into the underlying physics of energy deposition from short-pulse lasers in KDP and DKDP crystals.

The second objective of this proposal was to explore the mechanisms by which energy is deposited during damage in SiO_2 . We have measured how laser parameters such as pulse duration, pulse shape, wavelength, and fluence are relevant to energy deposition, for initiation and growth on the surface of SiO_2 optics.

Introduction

Potassium dihydrogen phosphate (KH₂PO₄) known as KDP and its deuterated analog DKDP, are commonly used as Second Harmonic Generators (SHGs) and Third Harmonic Generators (THGs) respectively. KDP is uniquely suitable for use in large aperture lasers because of its optical properties (birefringence & nonlinear index) and the rate at which large (300 kg) crystals may be grown (as fast as tens of mm a day). As with all optical materials KDP is susceptible to laser-induced damage both in its bulk and on its surface. KDP is somewhat unique in that bulk damage rather than surface damage is generally believed to be the limiting factor in operation.

Bulk damage in KDP is believed to be caused by small (~100 nm) defect clusters formed in the crystal during growth. The precise nature of these so-called "precursors" is not known but their existence is inferred from the nature of laser-induced bulk damage in KDP. Bulk damage in KDP manifests itself as small spherical micro-cavities surrounded by compacted material referred to as "pinpoints." Although, these pinpoints, once formed, have a tendency not to grow upon subsequent exposure to laser irradiation at fluences below ~20 J/cm², they can affect laser beam quality through scattering. Also, when pinpoints are formed near the surface of an optic, they can erupt on to the surface forming a large scattering site with the potential to grow.

The effect of scattered light on a laser beam can be described in terms of beam contrast, the ratio of RMS intensity fluctuations to average beam intensity. Although the point at which beam contrast becomes detrimental is somewhat application dependent, the larger the contrast in a laser beam, the lower the average fluence the laser must be operated to prevent further damage to optical components. The effect of damage-induced scatter contrast can be estimated from

$$C_f = \sqrt{C_i^2 + C_+^2}$$
, where $C_+ = \sqrt{2 \cdot f_s}$.

where C_f , C_i , C_+ and f_s are final contrast, initial contrast, contrast due to damage alone, and the fraction of light energy scattered by damage, respectively. Higher operating fluences will produce more pinpoints and longer pulse durations will produce larger pinpoints. As the total scatter produced by the pinpoints is dependent on both their number and size, it is important to understand the dependence of pinpoint size, and number density on the operational parameters of the testing laser.

It has been well established that post fabrication processing (or pretreating) can reduce the susceptibility of KDP to laser-induced bulk damage This phenomena is collectively known as conditioning. It is believed that the conditioning process in someway modifies the absorption characteristics of the damage precursors formed during crystal growth. Both thermal annealing and laser irradiation have produced a "conditioning" effect in KDP as determined by changes in the material response to either S/1 or $\rho(\phi)$ (or both) damage testing.

Thermal annealing of crystals to \sim 100 deg C produces a moderate enhancement to the damage resistance from 1ω light. However, enhancement to damage resistance from 2ω light is minimal at best, and undetectable for 3ω light.

The approach used for laser conditioning consists of pre-exposing a crystal to gradually increasing fluences with the objective of enhancing damage resistance. The effectiveness of laser conditioning is a function of the operating parameters of the conditioning laser (in addition to the operating parameters of the testing laser). The laser parameters important to conditioning are wavelength, pulse duration, peak fluence, pulse shape, and fluence step size. It has historically been accepted that 3ω light will provide the best conditioning for any test wavelength (with all other laser parameters fixed).

Earlier efforts at laser conditioning utilized long pulse (~10 to 20 ns) lasers because of their availability and high average power output. Recently, during our study of the effect of pulse duration and peak fluence (at constant wavelength and pulse shape, Gaussian-3ω pulse), we observed that the conditioning effect appears to be greatest for pulses with durations between 300 and 900 ps when damage tested with 3-ns 3ω light (see figure 1). During this study the peak fluences examined for pulse durations less than 1 ns were not held constant because they were limited by the power output of the conditioning laser. On the other hand, for pulse durations longer than 1 ns, the laser was capable of sufficient power output to produce peak fluences that would cause the damage to be dominated by surface rather than bulk effects. This distinction is important because at longer pulse lengths, the experiment was limited by the damage probability due the surface finish of the material (surface damage). At the short pulses no such limitation was reached, suggesting a further improvement in conditioning effect with short pulses is possible. This is in agreement with other experiments that have shown that higher fluences (for any given set of fixed conditioning laser parameters) produce better conditioning.

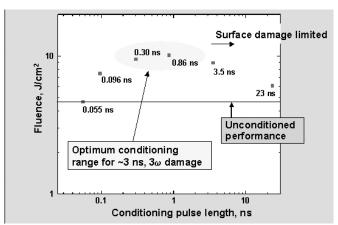


Figure 1. Plot of 3ω , 2.6 ns test fluences corresponding to a set scatter level for conditioning pulse lengths of 0.055 ns, 0.096 ns, 0.30 ns, 0.86 ns, 3.5 ns, and 23 ns. The optimal pulse length range for conditioning for 3ω , 2.6 ns damage appears to be 0.3 – 1 ns. For both the 0.30 ns and 0.86 ns conditioning was limited by the output of the conditioning laser, not any property of the sample.

Although no data for conditioning with different temporal pulse shapes exist, flat-in-time pulses are expected to produce a conditioning effect at lower fluences because flat-in-time pulses have been demonstrated to produce damage at lower fluences than Gaussian pulses.

Once the optimum parameters of the conditioning laser are established, it is still necessary to determine the procedure or protocol that will produce the maximum benefit. Past R/1 experiments conducted with 8 ns pulses have shown that small conditioning steps (i.e. the change in fluence from step to step of the order of 0.1-0.2 J/cm²) provide a maximum effective for conditioning. Although it is well known that larger conditioning step sizes will also provide effective conditioning, we have recently shown that there is a finite range in conditioning step sizes that provide the maximum conditioning effect. Preliminary tests with short pulse conditioning suggest that the optimal conditioning step size will be pulse length dependant.

The preliminary short pulse conditioning data discussed above was acquired in relatively small volumes with single shot laser experiments. Developing techniques applicable to conditioning large optics required that a small beam raster system be developed and optimized. As no commercial laser existed with the required power output over the range of pulse lengths needed, we designed and built one.

Unlike KDP, SiO₂ will generally not damage in the bulk. The areas of interest for fused silica are the initiation and growth of surface damage sites. A great deal of data has been generated in the past regarding these surface sites, in particular, important insights were gained in a previous LDRD which studied the interaction of 1ω , 2ω and 3ω light with optical materials. In that LDRD, we learned that all of our conceptions regarding how damage would scale from one wavelength to another, and how the presence of 1ω would impact damage at the drive wavelength were at best naïve. In this work we further study the effect of wavelength, pulse duration, and optic surface on damage growth and initiation.

Research Activities

The first objective of this proposal is to develop a fundamental understanding of how short-pulse (100 ps to 1 ns) laser parameters affect bulk conditioning and surface damage initiation in KDP and DKDP crystals. We will then use this information to develop off-line laser-based conditioning protocols that will significantly increase the damage resistance and extend the lifetime of frequency conversion crystals used in fusion-class laser systems. The second objective of this proposal is to explore the laser parameters important for energy deposition on fused silica optics. In pursuit of these objectives, we have divided our technical plan into a number of tasks. As our research progressed it became obvious that two of these tasks (4 and 6) were so interrelated that they should be merged. This will be discussed further in the task 4 & 6 accomplishments section.

- 1) Off-line determination of conditioning parameters of KDP
- 2) Construct of Prototype Conditioning Laser (PCL)
- 3) Use PCL to determine the optimal conditioning parameters for KDP and if SiO2 surfaces can be conditioned
- 4) Experimentally characterize the morphology of bulk and surface damage sites and determine their effects on contrast
- 5) Model and interpret bulk and surface damage
- 6) Characterize the effects of laser parameters on SiO2 damage initiation
- 7) Study the effects of laser parameters on Surface damage growth rate

Results and Technical outcome

Task 1 focused on narrowing the space of laser parameters relevant to effectively conditioning KDP and DKDP crystals. In this effort we developed new techniques to measure the densities of laser-induced damage $^{1,\,2}$. Using the new techniques we tested unconditioned KDP with pulse durations varying from 0.055 ns to 10 ns 3 . This testing indicated that the optimal conditioning pulse duration fell between 0.3 ns and 1 ns $^{3,\,4}$. Other conditioning parameters explored include wavelength, peak fluence, fluence step size, and number of conditioning pulses 5 . In addition we verified the previous measurements of how pulse duration effected damage density for 3ω light 3 as well as demonstrated that the relation changed for both 2ω light 4 and for 1ω light 6 .

Task 2 was to construct a prototype conditioning laser (PCL) suitable for determining the optimal conditioning recipe for large-aperture (40 cm) KDP and DKDP crystals 7 . The laser built met very demanding specifications: 38W output of 3ω light delivered in pulse of duration variable from 350 ps to 600 ps, a repetition rate of 300 Hz, and a spatially top hat beam with a contrast of less than 10%.

Task 3 was to use the PCL to determine the best laser parameters for conditioning KDP and DKDP crystals and explore the possibility of using the laser to treat SiO_2 optics. During this task we learned how to raster scan the PCL conditioning beam to condition large KDP Second Harmonic Generators⁸ (SHGs) and DKDP Third Harmonic Generators⁹ (THGs). We achieved improvements to the damage initiation characteristic of at least 250% for both optics types^{8, 9} as measured by both $\rho(\phi)$ and S/1 tests.

While achieving this core deliverable we also discovered that conditioning with the PCL can also shift the R/1 test, indicating that multiple pulses of various pulse durations may provide further conditioning of (D)KDP crystals. In addition to providing a future avenue of advance these results were crucial in guiding our modeling efforts which will be discussed in the task 5 accomplishments.

Because we elected to terminate the current project at 2.5 years the exploration of potential benefits of the PCL on SiO₂ will be moved to the out years and be further discussed in our exit strategy.

Task 4 was to characterize the effect of laser parameters on the morphology of damage sites in KDP and DKDP. As we gained knowledge from our work on this task and task 6, which was to explore the effect of the same laser parameters on initiation on the surface of SiO₂, it became obvious that the two very different materials had remarkably similar responses to most laser parameters. This was surprising because the two materials differ in composition, atomic structure, fabrication technique, mechanical strength, thermal properties, and potential impurities.

Despite the fact that KDP and SiO₂ differ in almost every way that seems relevant we found that the size and density of damage sites had qualitatively similar behavior as a function of fluence, pulse duration, and wavelength^{1, 6, 10-15}. The morphologies of the damage sites in both materials are quite complex including regions that have clearly been melted as well as more subtly modified areas^{11, 13}. In KDP where the damage primarily occurs in the bulk the damage sites manifest as small micro-cavities of a few microns in diameter. We have

shown that theses cavities consist of three main regions, a ruble filled core, and a thin shell of decomposed material both of which are encapsulated in a much larger region that is slightly densified, but still stoichiometric KDP. We have studied the decomposition of KDP under high pressures in diamond anvil experiments and observed similar results to those occurring during laser induced damage¹⁶.

Since SiO₂ primarily damages on the exit surface, rather than in the bulk, the gross geometries of the KDP and SiO₂ damage sites are quite different. The SiO₂ damage sites appear to be craters with melted cores which show considerable evidence that significant amounts of material was ejected in the forms of both chips and molten globules.

Task 5 was to model and interpret our observations of how laser parameters effect initiation, growth and conditioning. Early in this work the thermal absorption model was extended to include precursors shaped like plates and rods in addition to the balls used in the original model ^{17, 18}. Code designed to simulate the effect of precursor sizes and shapes on laser damage was written and used to evaluate the expanded thermal model against experimental observations ¹⁸. The thermal model was also extended in an attempted to explain conditioning ¹⁷.

Task 7 was to explore the effects of wavelength and pulse duration on growth rate. We determined that wavelength can make a dramatic difference in growth rate for longer pulses 19 . Additional work on the effect of multiple simultaneous wavelengths will be explored in out years as will be discussed further in our exit strategy. It was found that both front surface and exit surface damage had approximately the same fluence threshold for growth with 3ω light, but that the growth rate differed substantially 20 . This difference can likely be attributed to the geometry of the plasma formation relative to the direction of propagation of the laser 21 . We also found that the depth to diameter ratio of sites growing on the exit surface remain approximately constant 21 .

Exit Plan

As described in Task 3 and its accompanying references, this work has resulted in the development of techniques for processing THGs and SHGs which increase the damage resistance by ~250%. In addition to this very practical outcome we have produced the 21 publications¹⁻²¹ listed in the reference section. Finally, this work has opened up many new avenues of investigation for the phenomena of laser induced damage and conditioning. We have determined that multiple pulses with different durations may provide superior conditioning to anything we have yet explored. It has also become evident though this work that the effects of multiple simultaneous wavelengths are important to understanding the interaction between laser light and wide gap dielectrics. Both of these discoveries and the potential to use the PCL to improve SiO₂ will be explored further in a new LDRD (LDRD 08ERD-054) funded for FY 08.

References / Publications

- UCRL-JRNL-211068, C. W. Carr, M. D. Feit, M. C. Nostrand, et al., Measurement Science & Technology 17, 1958 (2006).
- UCRL-PROC-236247, M. J. Matthews and M. D. Feit, "Effect of Random Clustering on Surface Damage Density Estimates", Laser-Induced Damage in Optical Materials **6720** (2007).
- UCRL-PROC-207946, J. J. Adams, C. W. Carr, M. D. Feit, et al., "Pulse length dependence of laser conditioning and bulk damage in KD2PO4", Laser-Induced Damage in Optical Materials **5647**, 265 (2004).
- ⁴ UCRL-PROC-216743, J. J. Adams, J. R. Bruere, M. Bolourchi, et al., "Wavelength and pulselength dependence of laser conditioning and bulk damage in double-cut KH₂PO₄", Laser-Induced Damage in Optical Materials **5991** (2005).
- UCRL_CONF-218739, P. DeMange, R. A. Negres, C. W. Carr, et al., Optics Express 14, 5313 (2006).
- UCRL-JRNL-232337, D. A. Cross, M. R. Braunstein, and C. W. Carr, "The effect of pulse duration on laser-induced damage by 1053-nm light in potassium dihydrogen phosphate crystals", Laser-Induced Damage in Optical Materials 6403 (2006).
- ⁷ UCRL-JRNL-226179, J. Honig, J. Halpin, D. Browning, et al., Applied Optics **46**, 3269 (2007).
- UCRL-PROC-236318, J. J. Adams, J. A. Jarboe, M. D. Feit, et al., "Comparison between S/1 and R/1 tests and damage density vs. fluence ($\rho(\phi)$) results for unconditioned and sub-nanosecond laser-conditioned KD₂PO₄ crystals", Laser-Induced Damage in Optical Materials **6720** (2007).
- UCRL-CONF-226376, J. J. Adams, J. A. Jarboe, C. W. Carr, et al., "Results of sub-nanosecond laser-conditioning of KD₂PO₄", Laser-Induced Damage in Optical Materials **6403** (2006).
- UCRL-JRNL-214793, C. W. Carr and J. M. Auerbach, Optics Letters **31**, 595 (2006).
- UCRL-JRNL-211938, C. W. Carr, M. D. Feit, M. A. Johnson, et al., Applied Physics Letters **89** (2006).
- UCRL-PROC-207879, C. W. Carr, M. D. Feit, and A. Rubenchick, "Effect on scattering of complex morphology of DKDP bulk damage sites", Laser-Induced Damage in Optical Materials **5647**, 532 (2004).
- UCRL-PROC-216929, C. W. Carr, M. D. Feit, A. M. Rubenchik, et al., "A Summary of Recent Damage-Initiation Experiments on KDP Crystals", Laser-Induced Damage in Optical Materials **5991** (2005).
- ¹⁴ UCRL-PROC-226457, C. W. Carr, M. J. Matthews, B. J.D., et al., "The effect of laser pulse duratio non laser-induced damage in KDP and SiO2", Laser-Induced Damage in Optical Materials: 2006 6403 (2007).
- UCRL-JRNL-224567, C. W. Carr, J. B. Trenholme, and M. L. Spaeth, Applied Physics Letters **90** (2007).
- UCRL-JRNL-211938, T. J. Norman, J. M. Zaug, and C. W. Carr, Chemistry of Materials 18, 3074 (2006).
- UCRL-PROC-216930, M. D. Feit, A. M. Rubenchik, and J. B. Trenholme, "Simple model of laser damage initiation and conditioning in frequency conversion crystals", Laser-Induced Damage in Optical Materials **5991** (2005).
- UCRL-PROC-216931, J. B. Trenholme, A. M. Rubenchick, and M. D. Feit, "Size-Selection initiation model extended to include shape and random factors", Laser-Induced Damage in Optical Materials **5991** (2005).
- UCRL-PROC-216742, M. A. Norton, E. E. Donohue, M. D. Feit, et al., "Growth of Laser Damage in SiO2 under Multiple Wavelength Irradiation", Laser-Induced Damage in Optical Materials **5991** (2005).
- UCRL-CONF-226318, M. A. Norton, E. E. Donhue, M. D. Feit, et al., "Growth of Laser Damage on the Input Surface of SiO2 and 351 nm", Laser-Induced Damage in Optical Materials **6403** (2006).
- UCRL-PROC-236269, M. A. Norton, J. J. Adams, C. W. Carr, et al., "Growth of laser damage in fused silica: diameter to odepth ratio", Laser-Induced Damage in Optical Materials **6720** (2007).